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Published

With international search report.

(88) Date of publication of the international search report:

24 June 1999 (24.06.99)

(54) Title: COMPOSITE SUITABLE FOR USE IN ELECTROCHEMICAL CELLS

(57) Abstract

A composite comprises Aa) at least one first layer which comprises a mixture Ia, comprising a mix IIa consisting of a) from 1 to 95 % by weight of a solid III, preferably a basic solid III, having a primary particle size of from 5 nm to 20 µm and b) from 5 to 99 % by weight of a polymeric composition IV obtainable by polymerization of b1) from 5 to 100 % by weight, based on the composition IV, of a condensation product V of α) at least one compound VI which is able to react with a carboxylic acid or a sulfonic acid or a derivative or a mixture of two or more thereof, and β) at least 1 mol per mol of the compound VI of a carboxylic acid or sulfonic acid VII which contains at least one free-radically polymerizable functional group, or a derivative thereof or a mixture of two or more thereof, and b2) from 0 to 95 % by weight, based on the composition IV, of a further compound VIII having a mean molecular weight (number average) of at least 5000 and polyether segments in the main chain or a side chain, where the proportion by weight of the mix IIa in the mixture Ia is from 1 to 100 % by weight, and the layer is free of an electron-conducting, electrochemically active compound, wherein the first layer or layers and the second layer or layers which comprises an electron-conducting, electrochemically active compound, and B) at least one second layer or layers are joined to one another by one of the two methods V1 or V2: V1) Lamination of the first layer or layers with the second layer or layers under the action of heat or pressure or under the action of heat and pressure, or V2 Corona treatment of the first layer or layers, the second layer or layers or the first layer or layers and the second layer or layers and subsequent bringing together of the corona-treated first layer or layers with the corona-treated or untreated second layer or layers.

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## INTERNATIONAL SEARCH REPORT

International application No.

PCT/EP 98/06394

## A. CLASSIFICATION OF SUBJECT MATTER

IPC6: H01M 10/40, H01M 10/04

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC6: H01M

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WO 9601506 A1 (MOTOROLA INC.), 18 January 1996 (18.01.96), page 3, line 24 - page 4, line 15 --	1,2,10-12
X	DE 19612769 A1 (BASF AG), 2 October 1997 (02.10.97), page 1, line 1 - page 5, line 9 --	1,2
P,X	WO 9737397 A1 (BASF AKTIENGESELLSCHAFT), 9 October 1997 (09.10.97), page 4, line 15 - page 24, line 9 --	1,2,10-12
P,X	DE 19713072 A1 (BASF AG), 1 October 1998 (01.10.98), page 3, line 28 - page 10, line 5 --	1,2,10-12

☒ Further documents are listed in the continuation of Box C.☒ See patent family annex.

\* Special categories of cited documents:

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document but published on or after the international filing date

"I" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"(O)" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance: the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

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"&amp;" document member of the same patent family

Date of the actual completion of the international search

18 February 1999

Date of mailing of the international search report

15 April 1999 (15.04.99)

Name and mailing address of the ISA,

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ULLA GRANLUND

## INTERNATIONAL SEARCH REPORT

International application No.

PCT/EP 98/06394

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
E	WO 9844576 A1 (BASF AKTIENGESELLSCHAFT), 8 October 1998 (08.10.98), page 1, line 6 - page 38, line 30  --	1,2,10,12
A	DE 3920129 A1 (RICOH CO., LTD.), 28 December 1989 (28.12.89), page 2, line 39 - page 5, line 55; page 6, line 29 - line 42  -- -----	1-12

# INTERNATIONAL SEARCH REPORT

SA 213925

Information on patent family members

02/02/99

International application No.

PCT/EP 98/06394

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# INTERNATIONALER RECHERCHENBERICHT

Angaben zu Veröffentlichungen, die zur selben Patentfamilie gehören

ationales Aktenzeichen

PCT/EP 98/01763

Im Recherchenbericht angeführtes Patentdokument	Datum der Veröffentlichung	Mitglied(er) der Patentfamilie	Datum der Veröffentlichung
US 5348824 A	20-09-1994	CA 2109246 A	27-04-1995
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# INTERNATIONALER RECHERCHENBERICHT

Angaben zu Veröffentlichungen... die zur selben Patentfamilie gehören

Internationale Aktenzeichen  
PCT/EP 97/01594

Im Recherchenbericht angeführtes Patentdokument	Datum der Veröffentlichung	Mitglied(er) der Patentfamilie	Datum der Veröffentlichung
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## PATENT COOPERATION TREATY

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## NOTIFICATION OF ELECTION

(PCT Rule 61.2)

From the INTERNATIONAL BUREAU

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in its capacity as elected Office

Date of mailing (day/month/year)  
23 June 1999 (23.06.99)

International application No.  
PCT/EP98/06394

Applicant's or agent's file reference  
NAE19970565PC

International filing date (day/month/year)  
08 October 1998 (08.10.98)

Priority date (day/month/year)  
09 October 1997 (09.10.97)

Applicant

BAUER, Stephan et al

1. The designated Office is hereby notified of its election made:

☒ in the demand filed with the International Preliminary Examining Authority on:  
28 April 1999 (28.04.99)

☐ in a notice effecting later election filed with the International Bureau on:

2. The election ☒ was

☐ was not

made before the expiration of 19 months from the priority date or, where Rule 32 applies, within the time limit under Rule 32.2(b).

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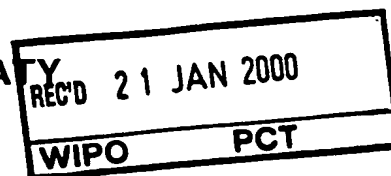
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ART 34 AMDT.

PATENT COOPERATION TREATY

PCT



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INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Article 36 and Rule 70)

Applicant's or agent's file reference NAE19970565P	<b>FOR FURTHER ACTION</b> See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416)	
International application No. PCT/EP98/06394	International filing date (day/month/year) 08/10/1998	Priority date (day/month/year) 09/10/1997
International Patent Classification (IPC) or national classification and IPC H01M10/40		
Applicant BASF AKTIENGESELLSCHAFT		

1. This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.



2. This REPORT consists of a total of 4 sheets, including this cover sheet.

- ☒ This report is also accompanied by ANNEXES, i.e. sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).

These annexes consist of a total of 6 sheets.

3. This report contains indications relating to the following items:

- I ☒ Basis of the report
- II ☐ Priority
- III ☐ Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
- IV ☐ Lack of unity of invention
- V ☒ Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
- VI ☐ Certain documents cited
- VII ☐ Certain defects in the international application
- VIII ☐ Certain observations on the international application

Date of submission of the demand 28/04/1999	Date of completion of this report 08/10/1998
Name and mailing address of the international preliminary examining authority:  European Patent Office D-80298 Munich Tel. +49 89 2399 - 0 Tx: 523656 epmu d Fax: +49 89 2399 - 4465	Authorized officer Del Piero, G Telephone No. +49 89 2399 8579 

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# INTERNATIONAL PRELIMINARY EXAMINATION REPORT

International application No. PCT/EP98/06394

## I. Basis of the report

1. This report has been drawn on the basis of (*substitute sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to the report since they do not contain amendments.*):

### Description, pages:

1-46 as originally filed

### Claims, No.:

1-12 as received on 17/12/1999 with letter of 17/12/1999

2. The amendments have resulted in the cancellation of:

- ☐ the description, pages:  
☐ the claims, Nos.:  
☐ the drawings, sheets:

3. ☐ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed (Rule 70.2(c)):

4. Additional observations, if necessary:

## V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

### 1. Statement

Novelty (N)	Yes:	Claims 1-12
	No:	Claims
Inventive step (IS)	Yes:	Claims 1-12
	No:	Claims
Industrial applicability (IA)	Yes:	Claims 1-12
	No:	Claims

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# INTERNATIONAL SEARCH REPORT

national Application No  
PCT/EP 98/01763

## C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	<p>US 5 514 461 A (MEGURO KAZUHIRO ET AL) 7  May 1996  see claims 1-9</p> <p>-----</p>	1-13

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**INTERNATIONAL PRELIMINARY  
EXAMINATION REPORT**

International application No. PCT/EP98/06394

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**2. Citations and explanations**

**see separate sheet**

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**INTERNATIONAL PRELIMINARY  
EXAMINATION REPORT - SEPARATE SHEET**

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International application No. PCT/EP98/06394

The present claims do not introduce matter extending beyond the content of the application as originally filed.

Although formulated in rather broad terms, the subject-matter of the current claims is not anticipated by any of the available citations, which either do not disclose the combination of all the ingredients of the composites according to the present claims or do not mention the combination of at least two layers forming the composites, as defined in the claims, by means of lamination under heat and/or pressure or corona treatment.

The provision of an alternative polymeric, filler-containing electrolyte composite system showing higher mechanical stability than conventional polymeric, filler-containing electrolytes under the stress conditions arising in the production of batteries can form the basis for the acknowledgment of an inventive step.

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We claim:

1. A composite comprising

Aa) at least one first layer which comprises a mixture Ia, comprising a mix IIa consisting of

a) from 1 to 95% by weight of a solid III, preferably a basic solid III, having a primary particle size of from 5 nm to 20  $\mu$ m and

b) from 5 to 99% by weight of a polymeric composition IV obtainable by polymerization of

b1) from 5 to 100% by weight, based on the composition IV, of a condensation product V of

$\alpha$ ) at least one compound VI which is able to react with a carboxylic acid or a sulfonic acid or a derivative or a mixture of two or more thereof, and

$\beta$ ) at least 1 mol per mol of the compound VI of a carboxylic acid or sulfonic acid VII which contains at least one free-radically polymerizable functional group, or a derivative thereof or a mixture of two or more thereof,

and

b2) from 0 to 95% by weight, based on the composition IV, of a further compound VIII having a mean

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molecular weight (number average) of at least 5000 and polyether segments in the main chain or a side chain,

where the proportion by weight of the mix IIa in the mixture Ia is from 1 to 100% by weight,

and the layer is free of an electron-conducting, electrochemically active compound,

and

B) at least one second layer which comprises an electron-conducting, electrochemically active compound,

wherein the first layer or layers and the second layer or layers are joined to one another by one of the two methods V1 or V2:

V1) Lamination of the first layer or layers with the second layer or layers under the action of heat or pressure or under the action of heat and pressure, or

V2) Corona treatment of the first layer or layers, the second layer or layers or the first layer or layers and the second layer or layers and subsequent bringing together of the corona-treated first layer or layers with the corona-treated or untreated second layer or layers.

## 2. A composite comprising

Ab) at least one first layer which comprises a mixture Ib comprising a mix IIb consisting of

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a) from 1 to 95% by weight of a solid III, preferably a basic solid, having a primary particle size of from 5 nm to 20  $\mu$ m and

b) from 5 to 99% by weight of a polymer IX obtainable by polymerization of

b1) from 5 to 75% by weight, based on the polymer IX, of a free-radically polymerizable compound X which is different from the carboxylic acid or the sulfonic acid VII or a derivative thereof, or a mixture of two or more thereof,

and

b2) from 25 to 95% by weight, based on the polymer IX, of a further compound VIII having a mean molecular weight (number average) of at least 5000 and polyether segments in the main chain or a side chain,

where the proportion by weight of the mix IIb in the mixture Ib is from 1 to 100% by weight

and the layer is free of an electron-conducting, electrochemically active compound,

and

B) at least one second layer which comprises an electron-conducting, electrochemically active compound,

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wherein the first layer or layers and the second layer or layers are joined to one another by one of the two methods V1 or V2:

5           V1)           Lamination of the first layer or layers with the second layer or layers under the action of heat or pressure or under the action of heat and pressure, or

10           V2)           Corona treatment of the first layer or layers, the second layer or layers or the first layer or layers and the second layer or layers and subsequent bringing together of the corona-treated first layer or layers with the corona-treated or untreated second layer or layers.

15           3.           A composite comprising at least one first layer Aa or at least one first layer Ab or at least one first layer Aa and at least one first layer Ab, at least one second layer B, each as defined in claim 1 or 2, and

20           C) at least one bonding layer.

25           4.           A composite as claimed in claim 3, wherein the bonding layer or layers C has/have a melting point which is lower than the melting point of the first layer or layers or the second layer or layers or the first and second layer or layers.

30           5.           A composite as claimed in claim 3 or 4, wherein the bonding layer or layers C is/are a polyethylene oxide, a polyvinyl ether, a polyacrylate, a polymethacrylate, polyvinylpyrrolidone, a polyurethane, a wax-like (co)polyolefin, a rubber-like material, polyisobutylene or a mixture of two or more thereof.

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6. A composite as claimed in any of claims 3 to 5, wherein the bonding layer or layers C comprise(s) a solid III, a plasticizer or a combination of two or more thereof.
- 5 7. A process for producing a composite as claimed in any of claims 1 to 6, which comprises joining the first layer or layers and the second layer or layers and, if present, the bonding layer or layers to one another by hot lamination.
- 10 8. A process for producing a composite as claimed in claim 1 or 2, which comprises subjecting the first layer or layers or the second layer or layers or the first layer or layers and the second layer or layers to a corona treatment and subsequently joining the first corona-treated layer or layers to the second corona-treated or untreated layer  
15 or layers.
9. A process for producing a composite as claimed in any of claims 3 to 6, which comprises applying at least one bonding layer to the first layer or layers, the second layer or layers or the first and second  
20 layer or layers and subsequently joining the first layer or layers to the second layer or layers via the bonding layer or layers.
10. The use of a composite as claimed in any of claims 1 to 6 for producing an electrochemical cell, in a sensor, an electrochromic  
25 window, a display, a capacitor or an ion-conducting film.
11. An electrochemical cell comprising a composite as claimed in any of claims 1 to 6 or a combination of two or more thereof.
- 30 12. The use of the electrochemical cell as claimed in claim 11 as an automobile battery, instrument battery, planar battery or polymer

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battery.

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## PCT

## INTERNATIONAL SEARCH REPORT

(PCT Article 18 and Rules 43 and 44)

Applicant's or agent's file reference <b>NAE19970565PC</b>	<b>FOR FURTHER ACTION</b> see Notification of Transmittal of International Search Report (Form PCT/ISA/220) as well as, where applicable, item 5 below.	
International application No. <b>PCT/EP 98/ 06394</b>	International filing date (day/month/year) <b>08/10/1998</b>	(Earliest) Priority Date (day/month/year) <b>09/10/1997</b>
Applicant <b>BASF AKTIENGESELLSCHAFT et al.</b>		

This International Search Report has been prepared by this International Searching Authority and is transmitted to the applicant according to Article 18. A copy is being transmitted to the International Bureau.

This International Search Report consists of a total of 3 sheets.



It is also accompanied by a copy of each prior art document cited in this report.

## 1. Basis of the report

- a. With regard to the **language**, the international search was carried out on the basis of the international application in the language in which it was filed, unless otherwise indicated under this item.



the international search was carried out on the basis of a translation of the international application furnished to this Authority (Rule 23.1(b)).

- b. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, the international search was carried out on the basis of the sequence listing :



contained in the international application in written form.



filed together with the international application in computer readable form.



furnished subsequently to this Authority in written form.



furnished subsequently to this Authority in computer readable form.



the statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.



the statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished

2. ☐ **Certain claims were found unsearchable** (See Box I).

3. ☐ **Unity of invention is lacking** (see Box II).

4. With regard to the **title**,

the text is approved as submitted by the applicant.



the text has been established by this Authority to read as follows:

5. With regard to the **abstract**,

the text is approved as submitted by the applicant.



the text has been established, according to Rule 38.2(b), by this Authority as it appears in Box III. The applicant may, within one month from the date of mailing of this international search report, submit comments to this Authority.

## 6. The figure of the drawings to be published with the abstract is Figure No.



as suggested by the applicant.



because the applicant failed to suggest a figure.



because this figure better characterizes the invention.



None of the figures.

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PCT/EP 98/06394

## A. CLASSIFICATION OF SUBJECT MATTER

IPC6: H01M 10/40, H01M 10/04

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC6: H01M

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X ✓	WO 9601506 A1 (MOTOROLA INC.), 18 January 1996 (18.01.96), page 3, line 24 - page 4, line 15 --	1,2,10-12
X ✓	DE 19612769 A1 (BASF AG), 2 October 1997 (02.10.97), page 1, line 1 - page 5, line 9 --	1,2
P,X ✓	WO 9737397 A1 (BASF AKTIENGESELLSCHAFT), 9 October 1997 (09.10.97), page 4, line 15 - page 24, line 9 --	1,2,10-12
P,X ✓	DE 19713072 A1 (BASF AG), 1 October 1998 (01.10.98), page 3, line 28 - page 10, line 5 --	1,2,10-12

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Date of the actual completion of the international search

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## C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

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E ✓	WO 9844576 A1 (BASF AKTIENGESELLSCHAFT), 8 October 1998 (08.10.98), page 1, line 6 - page 38, line 30  --	1,2,10,12
A ✓	DE 3920129 A1 (RICOH CO., LTD.), 28 December 1989 (28.12.89), page 2, line 39 - page 5, line 55; page 6, line 29 - line 42  -- -----	1-12

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**INTERNATIONAL SEARCH REPORT**  
Information on patent family members

213925

International application No.

02/02/99

PCT/EP 98/06394

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## INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

<b>(51) International Patent Classification</b> <sup>6</sup> : <b>H01M 10/40, 10/04</b>	<b>A3</b>	<b>(11) International Publication Number:</b> <b>WO 99/19917</b> <b>(43) International Publication Date:</b> 22 April 1999 (22.04.99)
<b>(21) International Application Number:</b> PCT/EP98/06394 <b>(22) International Filing Date:</b> 8 October 1998 (08.10.98) <b>(30) Priority Data:</b> 197 44 660.4      9 October 1997 (09.10.97)      DE <b>(71) Applicants (for all designated States except US):</b> BASF AK-TIENGESELLSCHAFT [DE/DE]; D-67056 Ludwigshafen (DE). GS JAPAN STORAGE [JP/JP]; Battery Co., Ltd., 1 Inobaba-cho Nishinosho Kisshoin, Minami-ku, Kyoto 601 (JP). <b>(72) Inventors; and</b> <b>(75) Inventors/Applicants (for US only):</b> BAUER, Stephan [DE/DE]; Hauptstrasse 65a, D-67126 Hochdorf-Assenheim (DE). BRONSTERT, Bernd [DE/DE]; Zanderstrasse 35, D-67166 Otterstadt (DE). MÖHWALD, Helmut [DE/DE]; Markwardstrasse 16, D-76855 Annweiler (DE). STEPHAN, Oskar [DE/DE]; Kolpingstrasse 1, D-68766 Hockenheim (DE). TUKAMOTO, Hisashi [JP/JP]; 1, Inobanba-cho, Nishinosho, Kisshoin, Minami-ku (JP). <b>(74) Agent:</b> ISENBRUCK, Günter; Bardehle et al, Theodor-Heuss-Anlage 12, D-68165 Mannheim (DE).		<b>(81) Designated States:</b> AL, AU, BG, BR, BY, CA, CN, CZ, GE, HU, ID, IL, JP, KR, KZ, LT, LV, MX, NO, NZ, PL, RO, RU, SG, SI, SK, TR, UA, US, Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE).  <b>Published</b> <i>With international search report.</i>  <b>(88) Date of publication of the international search report:</b> 24 June 1999 (24.06.99)
<b>(54) Title:</b> COMPOSITE SUITABLE FOR USE IN ELECTROCHEMICAL CELLS  <b>(57) Abstract</b>  A composite comprises Aa) at least one first layer which comprises a mixture Ia, comprising a mix IIa consisting of a) from 1 to 95 % by weight of a solid III, preferably a basic solid III, having a primary particle size of from 5 nm to 20 µm and b) from 5 to 99 % by weight of a polymeric composition IV obtainable by polymerization of b1) from 5 to 100 % by weight, based on the composition IV, of a condensation product V of α) at least one compound VI which is able to react with a carboxylic acid or a sulfonic acid or a derivative or a mixture of two or more thereof, and β) at least 1 mol per mol of the compound VI of a carboxylic acid or sulfonic acid VII which contains at least one free-radically polymerizable functional group, or a derivative thereof or a mixture of two or more thereof, and b2) from 0 to 95 % by weight, based on the composition IV, of a further compound VIII having a mean molecular weight (number average) of at least 5000 and polyether segments in the main chain or a side chain, where the proportion by weight of the mix IIa in the mixture Ia is from 1 to 100 % by weight, and the layer is free of an electron-conducting, electrochemically active compound, and B) at least one second layer which comprises an electron-conducting, electrochemically active compound, wherein the first layer or layers and the second layer or layers are joined to one another by one of the two methods V1 or V2: V1) Lamination of the first layer or layers with the second layer or layers under the action of heat or pressure or under the action of heat and pressure, or V2 Corona treatment of the first layer or layers, the second layer or layers or the first layer or layers and the second layer or layers and subsequent bringing together of the corona-treated first layer or layers with the corona-treated or untreated second layer or layers.		

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International application No.

PCT/EP 98/06394

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Date of the actual completion of the international search

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15 April 1999 (15.04.99)

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## INTERNATIONAL SEARCH REPORT

International application No.

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A	DE 3920129 A1 (RICOH CO., LTD.), 28 December 1989 (28.12.89), page 2, line 39 - page 5, line 55; page 6, line 29 - line 42  -- -----	1-12

# INTERNATIONAL SEARCH REPORT

SA 213925

Information on patent family members

02/02/99

International application No.

PCT/EP 98/06394

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